

## *Rapid Communication*

# Laser Photoionization Detection of the Rare Isotope $^3\text{He}$

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**Abstract.** Based on combination of the two-step collinear isotope-selective photoionization and time-of-flight separation of atoms in a fast modulated beam, a new technique has been developed for detecting the  $^3\text{He}$  rare isotope with an isotopic selectivity of up to  $10^{10}$ . The technique helped to detect optically, for the first time,  $^3\text{He}$  at a relative abundance of  $4 \cdot 10^{-8}$ .

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The solution of many problems in nuclear physics, geochemistry, oceanology, environmental protection, historical dating, etc., requires determination of ultralow number densities (less than  $10^{-9}$ ) of rare isotopes occurring amidst the abundant main isotopes of the same element. At present, this purpose is served, apart from the nuclear methods for measuring ultralow levels of radioactivity, by the accelerator mass spectrometry technique [1]. Many investigators have suggested that isotope-selective excitation by laser radiation should be used to develop a rare-isotope detector. A review of these proposals can be found in [2]. However, the maximum selectivities attained by means of laser techniques without preliminary concentration of the isotope to be detected range between  $10^5$  and  $10^7$  [3–6]. By using a preliminary three-stage enrichment, Thonnard and coworkers [7] have detected krypton isotopes with a natural relative abundance of  $10^{-12}$ . These results are substantially inferior to those obtained with the aid of accelerator mass spectrometry.

The present paper reports on the direct optical detection of the rare isotope  $^3\text{He}$  with a relative concentration of up to  $10^{-8}$  by the collinear laser photoionization of atoms in a fast modulated beam.

The idea of the collinear laser photoionization of rare isotopes, suggested in [8] and experi-

mentally tested in the experiments with potassium isotopes [4], consists in the isotope-selective excitation of the isotopic atoms of interest to a Rydberg state in a beam of fast atoms obtained by neutralizing a beam of ions accelerated through a given potential difference of  $U_A$ . Collinear excitation in such a beam gives rise to an additional kinematic isotope shift in the absorption spectrum of any atomic transition in any element which is much in excess of the natural isotope shift. Due to the velocity bunching of the atoms in the beam, the absorption linewidth can be brought down to the radiative linewidth [9, 10]. Thus, this method is potentially capable of an extremely high selectivity in the laser excitation of rare isotopic atoms.

However, experiments with the  $^3\text{He}$  isotope, whose natural content in air is  $1.4 \cdot 10^{-6}$  relative to the  $^4\text{He}$  isotope, have shown that the detection selectivity provided by this method is limited to  $10^6$  because of the background noise due to two collisional processes [11]. First, collisions between fast  $^4\text{He}$  atoms and the residual gas in the chamber lead, although with a low probability, to the nonselective collisional excitation of the atoms to Rydberg states which then undergo field ionization together with the  $^3\text{He}$  atoms to be detected. Secondly, the fast  $^4\text{He}$  atoms can be ionized by collision with the residual gas molecules in the region of field ionization of the Ryd-

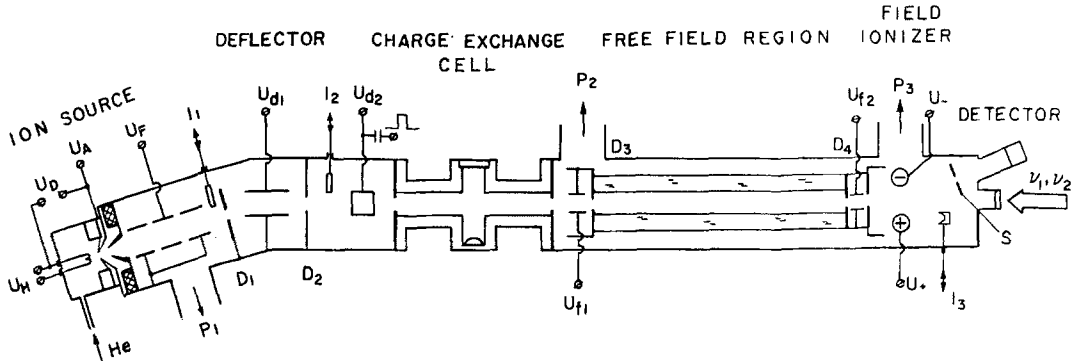


Fig.1. Schematic of the experimental setup. ( $D_1$ - $D_4$ : diaphragms;  $d_1$  and  $d_2$ : deflecting capacitors;  $f_1$  and  $f_2$ : filter capacitors;  $I_1$ - $I_3$ : atomic beam current meters; S: adjustable slit)

berg  $^3\text{He}$  atoms. To suppress this background noise, the method can be used in conjunction with various mass spectrometry techniques. In the present work, this was achieved through an additional time-of-flight separation of  $^4\text{He}$  and  $^3\text{He}$ , for which purpose use was made of an additional pulse intensity modulation of the continuous ion beam. This made it possible to reduce the background noise due to  $^4\text{He}^+$  ions by a factor of  $10^4$  and to detect  $^3\text{He}$  with a relative abundance as low as  $10^{-8}$ . This limit was set by the number of the  $^3\text{He}$  atoms excited by the low-repetition-rate tunable dye lasers used in the experiment and not by the background noise.

The experimental setup (Fig.1) was described in detail in [11]. The continuous beam of helium ions was produced by a hot-cathode gas-discharge source. The ion energy (3.9 keV) was determined by the extracting electrode potential  $U_A$ . The ion beam was collimated by a single lens and then deflected by the deflector  $d_1$  into a charge-exchange cell through the modulator  $d_2$ , which completely deflected the beam off axis when a dc voltage of  $U_{d_2} = 50$  V was applied to it.

To effect the additional time-of-flight separation of  $^3\text{He}^+$  and  $^4\text{He}^+$  ions, an electric pulse was applied to the modulator  $d_2$  so as to make it let the ions pass through for a short time of  $\tau_p = 0.5$   $\mu\text{s}$ . For the  $^3\text{He}$  and  $^4\text{He}$  ions to be completely separated by the time of arrival at the detector, the ion pulse duration must not exceed the difference between the times it takes for these ions to cover the distance  $L_1$  from the modulator  $d_2$  to the detector:

$$\tau = L_1 \left[ (M_{^3\text{He}})^{1/2} - (M_{^4\text{He}})^{1/2} \right] (2eU_A)^{-1/2}.$$

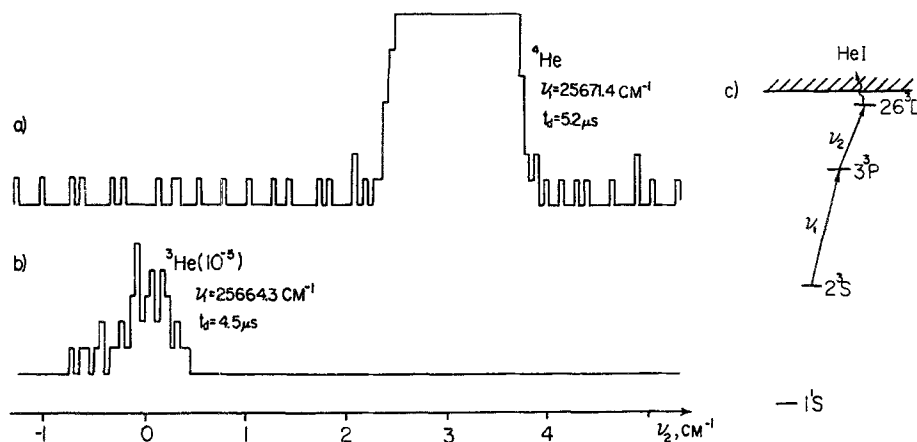
For ions with 3.9 keV energy,  $\tau = 0.7$   $\mu\text{s}$ .

In the charge-exchange cell containing potassium vapor, 40% of the ions were converted to metastable helium atoms in the  $2^1\text{S}$  and  $2^3\text{S}$  states, 3/4 of the atoms being formed in the  $2^3\text{S}$  triplet state [12]. The ions that failed to exchange their charge and the atoms excited in the charge-exchange cell to levels with principal quantum number  $n > 17$  were extracted from the beam by a filter capacitor with an applied potential of  $U_{f_1}$  (the capacitor field strength was 9 kV/cm).

At the moment the  $^3\text{He}$  atoms arrived at the exit from the field-free region (the diaphragm  $D_4$  was 2 mm in diameter) all the atoms in this region in the metastable  $2^3\text{S}$  state were excited in an isotope-selective fashion by means of two pulsed dye lasers 25 Hz in pulse repetition rate pumped by a XeCl laser 8 ns in pulse duration. The two laser beams were respectively collinear with, and counter to, the atomic beam.

Note that the trigger pulse applied to the modulator  $d_2$  could be repeated a time  $2\tau$  later, so that two bunches of  $^3\text{He}$  atoms spaced 70 cm apart would be in the 110 cm long field-free region at the moment the exciting laser pulse arrives. Such an atomic beam modulation would make it possible to double the sensitivity of the technique. Unfortunately, no such modulation was realized in the present work, there being no suitable strobed pulse registration system available,

The dye laser frequencies  $\nu_1$  and  $\nu_2$  were tuned precisely to the frequencies of the transitions  $2^3\text{S} \rightarrow 2^3\text{P}$  ( $\lambda_1 = 3888 \text{ \AA}$ ) and  $2^3\text{P} \rightarrow 26^3\text{D}$  ( $\lambda_2 = 7945 \text{ \AA}$ ) of the atoms in the fast beam (Fig. 2c), and so the  $^3\text{He}$  atoms were excited from the  $2^3\text{S}$  state to the Rydberg state  $26^3\text{D}$ . The laser fluences were high enough to saturate both transitions. In accordance with the statistical weights of the levels used, 5/9 of all the  $^3\text{He}$  atoms in the  $2^3\text{S}$  state were excited.



**Fig.2.** Ion signal as a function of the second-step excitation laser frequency  $\nu_2$ . (a) The first-step laser frequency is in resonance with  $^4\text{He}$ . The counting system gate is set to detect  $^4\text{He}$  ( $t_d = 5.2 \mu\text{s}$ ). Averaged over 64 pulses. (b) The first-step laser frequency is in resonance with  $^3\text{He}$ . The counting system gate is set to detect  $^3\text{He}$  ( $t_d = 4.5 \mu\text{s}$ ). Averaged over 64 pulses. (c) Diagram of two-step excitation of the He atom from the  $2^3\text{S}$  metastable state

The bandwidths of each laser pulse ( $\Delta\nu_{1,2} = 0.5 \text{ cm}^{-1}$ ) were substantially less than the isotope shifts in the first and second excitation steps ( $\delta\nu_{1,\text{is}} = 7.1 \text{ cm}^{-1}$  and  $\delta\nu_{2,\text{is}} = 3.2 \text{ cm}^{-1}$ ). The final spectral selectivity of excitation of the rare isotope  $^3\text{He}$  is related to absorption on the line wing of the main isotope  $^4\text{He}$  and is determined by the expression  $S_{1,2} = (\delta\nu_{1,2,\text{is}}/\Gamma)^2 (2\Gamma/\Delta\nu_{1,2})$ ,  $\Gamma$  being the absorption line half-width, provided that the condition  $\Gamma < \Delta\nu_{1,2} \ll \delta\nu_{1,2,\text{is}}$  is satisfied and the absorption line wing has a Lorentzian profile. For the helium atom transitions used,  $\Gamma = 0.84 \text{ MHz}$ . The excitation selectivity of  $^3\text{He}$  in the first and second excitation steps was  $7.2 \cdot 10^6$  and  $1.4 \cdot 10^6$ , respectively. The total spectral selectivity of the two-step excitation was  $S = S_1 S_2 = 10^{13}$ . The selectivity of multistep excitation has been considered in detail in [2, 13].

The Rydberg atoms were ionized by the electric field of an ionizer and deflected onto the detector. The detection of the  $^3\text{He}^+$  ions was effected by means of a gated counting system during a time interval of  $\tau_{\text{det}} = 0.6 \mu\text{s} < \tau$  delayed for a time of  $t_d = 4.5 \mu\text{s}$  relative to the modulating pulse applied to the electrode  $d_2$ . The background ions resulting from collisional processes involving the atoms of the main isotope  $^4\text{He}$  were delayed for  $5.2 \mu\text{s}$ .

To prepare calibration mixtures, use was made of 99% pure  $^3\text{He}$  and  $^4\text{He}$  containing less than  $10^{-7}$   $^3\text{He}$ . Use was also made of helium

extracted from atmospheric air with a relative  $^3\text{He}$  content of  $1.4 \cdot 10^{-6}$  [14]. This helium was obtained from a helium-neon mixture taken from a nitrogen liquefier and then purified with liquid helium.

Figures 2a and b illustrate the isotopic selectivity of the process of ionization of He atoms. Figure 2a shows the ion signal obtained while varying the frequency  $\nu_2$  of the second-step excitation laser, the first-step excitation laser frequency  $\nu_1 = 25671.4 \text{ cm}^{-1}$  being in resonance with  $^4\text{He}$  ( $t_d = 5.2 \mu\text{s}$ ). The background ions in the spectrum of  $^4\text{He}$  are due to the two collisional processes mentioned earlier. The ion signal from  $^3\text{He}$  with a relative content of  $10^{-5}$  is shown in Fig.2b. It was obtained with the frequency  $\nu_1$  shifted  $7.1 \text{ cm}^{-1}$  toward the "red" side and the counting system delay time  $t_d$  set at  $4.5 \mu\text{s}$ . The rate of counting background ions was so low that it could not be measured with the laser pulses recurring at a frequency of  $f = 25 \text{ Hz}$ , which corresponded to relative concentrations of  $^3\text{He}$  below  $10^{-8}$ .

Figure 3 shows the ion signal as a function of the relative content of the  $^3\text{He}$  isotope. The measured relative content of  $^3\text{He}$  in the helium gas used to prepare the calibration mixtures was  $4 \cdot 10^{-8}$ .

The sensitivity of the detection technique under consideration is limited by the number of  $^3\text{He}$  atoms excited in the beam. The rate of counting rare isotope atoms with a relative content of  $c$  is  $N = I \tau_p f \eta c$ , where  $I$  is the continuous atomic beam current (atoms/s),  $\tau_p$  the atomic beam current pulse duration,  $f$  the laser pulse repetition rate, and  $\eta$  the efficiency of exciting the atoms and detecting the ions produced. With  $f = 25 \text{ Hz}$ ,  $I = 10^{11} \text{ atoms/s}$ ,  $\tau_p = 0.5 \mu\text{s}$ ,  $\eta = 0.1$ , and  $c = 10^{-5}$ , the rate of counting  $^3\text{He}$  atoms

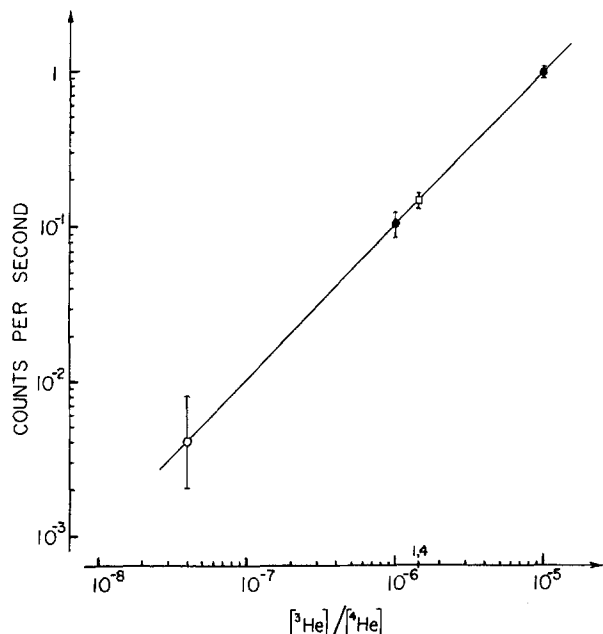


Fig.3. Ion signal from  $^3\text{He}$  ( $t_d=4.5\mu\text{s}$ ) as a function of the relative content  $[^3\text{He}]/[^4\text{He}]$ . (Full circles: calibration mixtures; open square: helium extracted from atmospheric air; open circle: helium used to prepare the calibration mixture)

will be  $N = 1.25$  counts/s. The sensitivity of the technique, defined as the number of counts per rare isotope atom in the beam,  $N/I_c$ , is equal to  $1.25 \cdot 10^{-6}$  counts/atom. This sensitivity can be substantially improved by using dye lasers pumped by a Cu-vapor laser with a higher pulse repetition frequency ( $f=10^4$  Hz) and modulating the atomic beams with a pair of trigger pulses as indicated earlier. In that case, the sensitivity of the technique will rise to  $10^{-3}$  counts/atom, which will correspond to a counting rate of 11 count/s at a  $^3\text{He}$  concentration of  $10^{-8}$ . We measured the background ion counting rate with the atomic beam modulated by single trigger pulses following at a rate of  $10^4$  Hz in the absence of laser radiation. This rate was  $0.5 \cdot 10^{-2}$  counts/s, which corresponded to a  $^3\text{He}$  concentration of  $10^{-10}$ .

Thus, the described technique for detecting the rare isotope  $^3\text{He}$ , based on combining the

collinear isotope-selective photoionization of fast atoms with the time-of-flight separation of isotopes, makes it possible to measure the relative content of  $^3\text{He}$  as low as  $10^{-10}$ . The determination of the isotopic composition of helium is essential to the solution of many problems in geochemistry and geophysics, cosmochemistry, oceanology, and so on [14]. The isotopic ratio  $[^3\text{He}]/[^4\text{He}]$  in various objects ranges between  $10^{-4}$  and  $10^{-10}$ . The method of collinear laser ionization of fast atoms can be used to solve these problems and to detect other rare isotopes as well.

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